

# Multi-Species Pair Annihilation Reactions

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We consider diffusion-limited reactions  $A_i + A_j \rightarrow \emptyset$  ( $1 \leq i < j \leq q$ ) in  $d$  space dimensions. For  $q > 2$  and  $d \geq 2$  we argue that the asymptotic density decay for such mutual annihilation processes with equal rates and initial densities is the same as for single-species pair annihilation  $A + A \rightarrow \emptyset$ . In  $d = 1$ , however, particle segregation occurs for all  $q < \infty$ . The total density decays according to a  $q$  dependent power law,  $\rho(t) \sim t^{-\alpha(q)}$ . Within a simplified version of the model  $\alpha(q) = (q - 1)/2q$  can be determined exactly. Our findings are supported through Monte Carlo simulations.

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A large variety of systems in physics, chemistry, biology, and ecology can be modeled in terms of diffusion-limited reactions. This is because of their unifying feature of being composed of mobile agents ('particles') which interact upon encounter. The traditional mean-field rate equations for such systems apply only to homogeneous particle densities. However, in many such systems the spatial dimension  $d$  has one or more critical values below which density fluctuations invalidate the rate equations and new phenomena appear. The fluctuations may stem from, *e.g.*, reaction-induced noise or initial state randomness, and typically dominate the system's large-scale, long-time behavior [1]. In order to extract the newly emerging nonequilibrium behavior, more sophisticated methods are needed: extensive numerical simulations (for recent overviews, see Ref. [2]) along with powerful analytical methods such as scaling approaches; mappings to field theories followed by renormalization group; and exact solutions (mostly limited to  $d = 1$ ) [3].

The theory of annihilation reactions has two landmarks, of which one, the single-species pair annihilation reaction  $A + A \rightarrow \emptyset$ , represents the simplest model case [4]. The other one, the two-species annihilation  $A + B \rightarrow \emptyset$ , is considerably more subtle. It exhibits the remarkable phenomenon that for  $d < 4$  and for an initially random particle distribution with equal densities  $\rho_A(0) = \rho_B(0)$ , the two species segregate into pure  $A$  and pure  $B$  domains, and the annihilations become localized within sharp reaction fronts between the domains [5]. It is a natural next step to ask for the long-time decay properties of a system of  $q$  species that mutually annihilate according to  $A_i + A_j \rightarrow \emptyset$ , with  $1 \leq i < j \leq q$  [6]. This 'mutual annihilation model' (MAM) is the subject of this Letter [7]. A special case is the fully symmetric MAM, characterized by equal reaction rates  $\lambda_{ij}$ , equal diffusion constants  $D_i$ , and equal initial densities  $\rho_i(0)$ . We will find that in  $d \geq 2$  for all  $2 < q < \infty$  the fully symmetric MAM behaves as the single-species pair annihilation process: the total density  $\rho(t) = \sum_i \rho_i(t) \sim t^{-1}$ . In stark contrast, in  $d = 1$  it exhibits species segregation, and is characterized by

$$\rho(t) \sim t^{-\alpha(q)} \quad (1)$$

with a  $q$  dependent exponent given, within the approach presented below, by  $\alpha(q) = (q - 1)/2q$ . We note that for  $q \rightarrow \infty$ , two particles of the same species meet with zero probability; the distinction between the different species then becomes irrelevant, and this model is equivalent to the  $A + A \rightarrow \emptyset$  reaction [8], with known  $\alpha(\infty) = 1/2$ .

In order to set the stage for our arguments, we briefly summarize the physics of the single- and two-species annihilation processes. For  $A + A \rightarrow \emptyset$  the mean-field rate equation  $\dot{\rho}_A = -\lambda\rho_A^2$  with the solution  $\rho_A(t) \sim 1/\lambda t$  provides a valid description only for  $d > 2$ . For  $d < 2$  nearby reactant pairs quickly annihilate, leaving only well-separated particles, which in turn slows the density decay down. These *anti*-correlations mimic a repulsion between the particles; in a field theory representation of the associated master equation [9] they lead to a downward renormalization of  $\lambda$ . As the diffusion propagator remains unchanged, the perturbation series is readily summed to all orders; one finds  $\rho_A(t) \sim t^{-d/2}$  for  $d < 2$  and  $\rho_A(t) \sim t^{-1} \ln t$  at the critical dimension  $d_c = 2$  [4].

For the two-species pair annihilation  $A + B \rightarrow \emptyset$  the rate equations read  $\dot{\rho}_{A/B} = -\lambda_{AB}\rho_A\rho_B$ . With equal initial densities  $\rho_A(0) = \rho_B(0)$  they are solved again by  $\rho_{A/B}(t) \sim 1/t$ ; with  $\rho_A(0) > \rho_B(0)$ , say, one obtains  $\rho_B(t) \sim \exp(-\lambda_{AB}[\rho_A(0) - \rho_B(0)]t)$  for the minority species, while the majority approaches a saturation density  $\rho_A(\infty)$ . In order to establish the effects of spatial fluctuations, it is crucial to notice that the density difference  $\rho_A - \rho_B$  remains conserved under the reactions; for  $D_A = D_B$  it simply obeys the diffusion equation [10]. Initial density difference fluctuations therefore amplify in time relative to the total density. As a consequence, when  $\rho_A(0) = \rho_B(0)$ , then for  $d < d_c = 4$  the system segregates into domains and the density decay is slowed down to  $\rho_{A/B}(t) \sim t^{-d/4}$  [11]. The renormalization group provides a firm basis [5] for these arguments, at least for  $2 \leq d < 4$ : An effective field theory can be derived that corresponds to the rate equations plus diffusion terms, which establishes the segregation.

For unequal diffusion constants,  $D_A \neq D_B$ , this picture is not qualitatively altered; however, special initial conditions may change it. Consider, *e.g.*, particles that initially alternate in one-dimensional space,  $ABAB\dots$ ,

and that upon encounter react with probability one. The distinction between  $A$  and  $B$  is then meaningless and the system is in the  $A + A \rightarrow \emptyset$  universality class [8]. For unbalanced initial conditions,  $\rho_A(0) > \rho_B(0)$ , stretched [12] rather than simple exponential relaxation ensues for  $d < 2$ :  $\ln \rho_B(t) \sim -t^{d/2}$ , whereas  $\ln \rho_B(t) \sim -t/\ln t$  at  $d_c = 2$  [11]. If the two species are already segregated initially,  $d_c = 2$  also is the sole critical dimension [13].

This summary helps us classify the possible scenarios for the  $q$ -species MAM with arbitrary parameters  $\lambda_{ij}$ ,  $D_i$ , and  $\rho_i(0)$ . Generically we expect that after some crossover time only the least reactive and initially most numerous species will have survived, resulting in an effective two-species problem. After this reduction of  $q$  to the effective value  $q_{\text{eff}} = 2$  the final asymptotic decay laws will be those of the two-species system with unequal initial densities discussed above. However, on special sub-manifolds of parameter space, and in particular in the presence of symmetries, reduction to  $q_{\text{eff}} = 2$  may not be possible and novel behavior may appear. That not *all* symmetries lead to new behavior may be illustrated by the cyclic reaction scheme  $A + B \rightarrow \emptyset$ ,  $B + C \rightarrow \emptyset$ ,  $C + D \rightarrow \emptyset$ , and  $D + A \rightarrow \emptyset$ , all with equal rates and initial densities. Here, we may readily identify the species  $A \equiv C$  and  $B \equiv D$ , respectively, which takes us back to the  $A + B \rightarrow \emptyset$  reaction with  $\rho_A(0) = \rho_B(0)$ . In this Letter we address the most prominent case that requires special consideration, and will in fact lead to novel effects, namely the fully symmetric MAM, in which all  $q$  species are equivalent (whence  $q_{\text{eff}} = q$ ).

First, we notice that the renormalization of the annihilation vertices in this  $q$ -species model is independent of  $q$  and identical to that of the  $A + A \rightarrow \emptyset$  reaction [5], with  $d_c = 2$  [4]. Second, for  $q > 2$  there exists no microscopic, local conservation law. As a consequence, following the arguments in Ref. [5], any memory of the initial state will eventually become lost. This eliminates the segregation mechanism at work in the  $q = 2$  case. Next we invoke the mean-field rate equations and conclude that  $\rho_i(t) = \rho(t)/q \sim 1/t$  for  $d > 2$  [14].

For  $d \leq 2$ , however, one needs to extract the correct asymptotic scaling from the Callan–Symanzik renormalization group equation. This requires an explicit computation of the density (a function of its initial value) as a power series in the renormalized annihilation rate  $\lambda_R$ . At the critical dimension  $d_c = 2$  the renormalized rate flows to zero logarithmically,  $\lambda_R(t) \sim 1/\ln t$ , leaving merely the tree diagram contributions that correspond to the solution of the rate equation. Thus we predict  $\rho_i(t) = \rho(t)/q \sim t^{-1} \ln t$  in  $d = 2$ . The difficulty for  $d < 2$  is to demonstrate that for large values of the relevant operator  $\rho(0)$  the power series remains properly controlled [4]. For  $d < 2$  this has proven elusive even for the two-species system [5]. Moreover, in one space dimension blockage effects of (hard-core) particles have been found to often play a crucial role in multi-

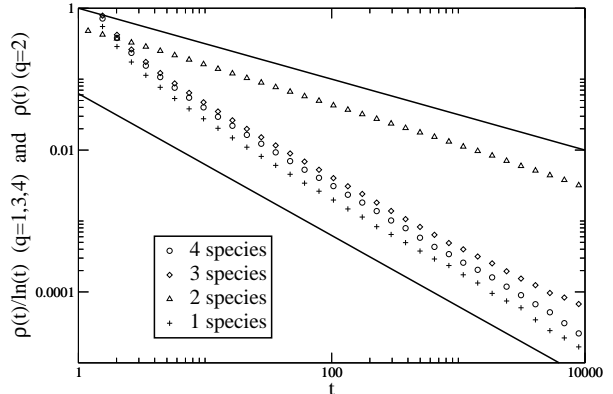


FIG. 1. Monte Carlo results for the total density decay vs. time in the pair annihilation reactions  $A + A \rightarrow \emptyset$  and  $A_i + A_j \rightarrow \emptyset$  with  $1 \leq i < j \leq q$  ( $q = 2, 3, 4$ ) for equal initial densities in two dimensions. The plots for  $q \neq 2$  depict  $\rho(t)/\ln t$ ; the solid lines indicate the functions  $t^{-1}$  and  $t^{-1/2}$ .

species reaction-diffusion processes [15]. We shall see that the  $q$ -species MAM, too, develops entirely novel features when restricted to a linear chain: The  $q$  species segregate into well-defined domains, which remain stable because of the mutual annihilation processes that prevent species mixing and the special one-dimensional topological constraints that do not allow a given species to interact with all others. As a consequence, we find that for all  $2 \leq q < \infty$  that the total density decays in  $d = 1$  according to the power law (1).

We now present our numerical evidence in  $d = 1$  and  $d = 2$ , and then proceed with the analysis of the one-dimensional model. In order to check the predicted universal decay law in two dimensions, we performed Monte Carlo simulations on a  $512 \times 512$  square lattice with hard-core particles. Starting from a full lattice with random distribution of  $q$  equally abundant species ( $q = 2, 3, 4$ ), we let the particles perform unbiased random walks and annihilate with probability one upon encounter with a different species. One Monte Carlo time step was considered complete after  $N(t)$  trials, with  $N(t)$  the number of remaining particles at that instant. The results, shown in Fig. 1, clearly support  $\rho(t) \sim t^{-1} \ln t$  for the  $q$ -species MAM with  $q = 3, 4$ ; this is similar to the decay law of the  $A + A \rightarrow \emptyset$  reaction, and in contradistinction to  $\rho(t) \sim t^{-1/2}$  for  $q = 2$ . We have also checked the purely mean-field behavior for  $q = 3, 4$  in a  $50^3$  cubic lattice [16].

In  $d = 1$ , however, simulations of the MAM with equal initial densities yield decay laws that differ importantly from both the  $A + A \rightarrow \emptyset$  and  $A + B \rightarrow \emptyset$  cases [8]. Figure 2 shows our Monte Carlo results for  $q = 2, 3, 4, 5$  on a chain of  $10^5$  sites with periodic boundary conditions. Evidently at long times  $\rho(t) \sim t^{-\alpha(q)}$ , where  $\alpha(q)$  increases with  $q$  from  $\alpha(2) = 1/4$  (the  $A + B \rightarrow \emptyset$  value) to  $\alpha(\infty) = 1/2$  (the  $A + A \rightarrow \emptyset$  value).

In order to study the one-dimensional MAM we simplify it such as to retain only its barest essentials. This

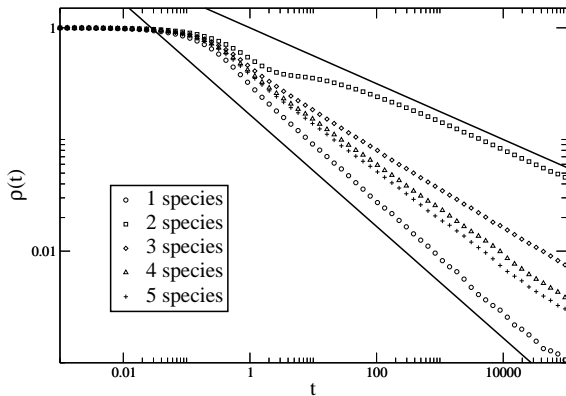


FIG. 2. Monte Carlo results for the total density decay  $\rho(t)$  vs.  $t$  in the pair annihilation reactions  $A + A \rightarrow \emptyset$  and  $A_i + A_j \rightarrow \emptyset$  with  $1 \leq i < j \leq q$  ( $q = 2, 3, 4, 5$ ) and equal initial densities in one dimension. The solid lines indicate the functions  $t^{-1/2}$  and  $t^{-1/4}$ .

simplified version, to be referred to as SMAM, arises from the following considerations. The one-dimensional system may at any time be decomposed into a sequence of domains, each containing only a single particle species. Owing to the diffusive nature of the process the typical domain size increases as  $L(t) \sim (Dt)^{1/2}$ . Let us assume the asymptotic decay law  $\rho(t) \sim \rho_0(\rho_0^2 Dt)^{-\alpha}$ , where  $\alpha$  remains to be determined. The average particle number in a domain then scales as  $\bar{n}(t) = L(t)\rho(t) \sim (\rho_0^2 Dt)^{-\alpha+1/2}$ , and phase segregation occurs only if  $\alpha < 1/2$ . Adjacent domains are separated by reaction zones, of which there are  $1/L(t)$  per unit of length. Therefore, as argued in Ref. [17] for the two-species case, the total particle density decreases as  $\dot{\rho}(t) = -\kappa(t)/L(t)$ , with  $\kappa(t)$  the typical number of annihilations per unit of time in a zone. The SMAM is now defined by the assertion that fluctuations in the annihilation rate  $\kappa(t)$ , whether in the course of time or between different reaction zones, are irrelevant and may be ignored; *i.e.*, the particle content of each domain, owing to the annihilations taking place at both of its ends, decreases at the uniform rate  $2\kappa(t)$ . To complete the picture, we need to specify what happens when a domain loses all its particles: Then, with probability  $1/(q-1)$ , the left and right neighboring domains contain identical species, and consequently fuse into a single new domain; or, with the complementary probability  $(q-2)/(q-1)$ , they contain different particle species and a new reaction zone appears.

The SMAM may be cast into an explicit algorithm. We consider a one-dimensional lattice whose sites  $1 \leq i \leq N^{(0)}$  represent the *domains* of the original MAM. We randomly select initial values  $n_i^{(0)}$  for the particle numbers in each domain. This random initial state evolves in time via *deterministic* iterations. The  $(k+1)$ th iteration changes the total number of sites from  $N^{(k)}$  to  $N^{(k+1)}$  and converts the integer set  $\{n_i^{(k)}\}_{i=1}^{N^{(k)}}$  to  $\{n_j^{(k+1)}\}_{j=1}^{N^{(k+1)}}$  by successive application of the following four iteration

steps: (i) All  $n_i^{(k)}$  are reduced by 1. (ii) All sites that as a result have become empty, are eliminated and the other sites are reconnected without reordering. (iii) Any two sites that as a result have become neighbors, are, with probability  $1/(q-1)$ , fused into a single site whose number variable is the sum of the number variables of the fusing sites. (iv) The remaining sites are relabeled with an index  $1 \leq j \leq N^{(k+1)}$ . The  $k$ th iteration yields the total number of domains  $N^{(k)}$  and the average particle number  $\bar{n}^{(k)}$  per domain. The particle density and the physical time follow from  $\rho^{(k)}/\rho^{(0)} = N^{(k)}\bar{n}^{(k)}/N^{(0)}\bar{n}^{(0)}$ , and  $t^{(k)}/t^{(0)} = [N^{(0)}/N^{(k)}]^2$ , as  $N(t) \sim L(t)^{-1} \sim t^{-1/2}$ .

A key feature of the SMAM is that at every iteration step  $k$  the numbers  $n_i^{(k)}$  are uncorrelated, for they descend from disjoint sets of ‘ancestor’ variables. Therefore this model obeys an exact closed set of equations, which we will now derive and analyze. Let us for the moment suppress the iteration superscript  $(k)$ , and denote, preceding the  $k$ th iteration, the total number of domains by  $N$ , the total number of domains containing  $n$  particles by  $M_n$ , and their relative abundance by  $f_n = M_n/N$ . Primed symbols will indicate the corresponding quantities after the  $k$ th iteration. In one iteration the total number  $N$  of domains diminishes by  $M_1$  due to step (i). Step (iii) results in the additional disappearance of domains; calculating their exact number requires taking into account all instances where two or more vanishing domains form a sequence of nearest or next-nearest neighbors. After some combinatorics one finds [16] that  $N'$  and  $N$  are related by  $N' = (1 - f_1)[1 - f_1/(q-1)]N$ . By means of more elaborate combinatorial analysis one may express the final number  $M'_n$  of  $n$  particle domains in terms of the initial  $M_m$ . The intensive variables  $f_n$  ( $n = 1, 2, \dots$ ) then obey the recursion relation

$$f'_n = [1 + (q-2)\tilde{f}_1](f_{n+1} + \tilde{f}_1\mathcal{R}_n), \quad (2)$$

with the abbreviations  $\tilde{f}_1 = f_1/[(q-1)(1-f_1)]$  and  $\mathcal{R}_n = \sum_{s=2}^{\infty} \tilde{f}_1^{s-2} \sum_{m_1, \dots, m_s=1}^{\infty} f_{m_1+1} \cdots f_{m_s+1} \delta_{n, m_1 + \dots + m_s}$ . The term of index  $s$  represents the creation of a domain of  $n$  particles by simultaneous fusion of  $s$  domains. The fusions with  $s \geq 3$  are clearly very model specific and one would expect the essential physics to be embodied already in the lowest-order nonlinearity. Indeed, by truncating Eq. (2) after the  $s = 2$  term one obtains an elegant Boltzmann-like equation; the mathematical analysis below is easier, however, if all terms are retained.

To find a solution to Eq. (2) we substitute an exponential distribution  $f_n = \epsilon(1-\epsilon)^{n-1}$ . The recursion then yields a similar distribution, but with a new parameter  $\epsilon'$  related to  $\epsilon$  by  $\epsilon' = \epsilon[1 - \epsilon/(q-1)]$ . For this solution  $f_1 = \epsilon$ , which may be substituted in the relation linking  $N'$  to  $N$ . Since  $\bar{n} = 1/\epsilon$ , the total particle density obeys  $\rho'/\rho = N'\epsilon/N\epsilon' = 1 - \epsilon$ . After restoring the iteration indices we obtain the pair of recursion relations

$$\epsilon^{(k+1)} = \epsilon^{(k)}[1 - \epsilon^{(k)}/(q-1)], \quad (3)$$

$$\rho^{(k+1)} = \rho^{(k)}[1 - \epsilon^{(k)}], \quad (4)$$

$$\ell(t) \sim t^{\lambda(q)} \quad (8)$$

to be solved with initial condition  $0 < \epsilon^{(0)} < 1$  [e.g., for a random initial distribution  $\epsilon^{(0)} = (q-1)/q$ ] and  $\rho^{(0)}$ . The solution of Eqs. (3) and (4) determines  $\rho^{(k)}$  and  $t^{(k)} = t^{(0)}[\rho^{(0)}\epsilon^{(0)}/\rho^{(k)}\epsilon^{(k)}]^2$ ; the desired function  $\rho(t)$  is then obtained by eliminating the index  $k$ .

We have been able to carry this through explicitly only in an asymptotic expansion for large  $k$ . Whereas its leading order is readily evaluated, more attention is required to deal with the subleading correction. By analyzing the recursion relation (3) one finds that

$$\epsilon^{(k)} = \frac{q-1}{k} \left[ 1 - \frac{\log ck}{k} + \mathcal{O}\left(\frac{\log^2 k}{k^2}\right) \right], \quad (5)$$

where  $c$  is a function of  $\epsilon^{(0)}/(q-1)$ . Analyzing Eq. (4) with (5) inserted then yields

$$\rho^{(k)} \simeq \frac{A\rho^{(0)}}{k^{q-1}} \left[ 1 - (q-1)\frac{\log ck - (q-2)/2}{k} \right] \quad (6)$$

with  $A = \lim_{k \rightarrow \infty} k^{q-1} \prod_{\ell=0}^{k-1} [1 - \epsilon^{(\ell)}]$ . Expressing  $t^{(k)}$  as a function of  $k$  and inverting leads to  $k(t) \simeq (Ct)^{1/2q} - [\log(Ct) + 2q \log c - (q-1)(q-2)]/2q$  with  $C = A^2(q-1)^2/\epsilon^{(0)2}t^{(0)}$ . Finally, upon substitution in Eq. (6),

$$\rho(t) \simeq A\rho^{(0)} \left[ (Ct)^{-\alpha(q)} + \frac{(q-1)(q-2)}{2q} (Ct)^{-1/2} \right] \quad (7)$$

with  $\alpha(q) = 1/2 - 1/2q < 1/2$ , confirming species segregation self-consistently and establishing Eq. (1) for the leading density decay of the SMAM. For  $q = 2$  we recover  $\alpha(2) = 1/4$ , and the limit  $q \rightarrow \infty$  gives correctly  $\alpha(\infty) = 1/2$ . Notice that the term with  $\log(Ct)$  and the dependence on  $c$  have canceled out in Eq. (7). The next-to-leading behavior is identical with the power law decay for the  $A + A \rightarrow \emptyset$  reaction in  $d = 1$ . Its relative amplitude increases with  $q$ ; thus it becomes numerically difficult to distinguish it from the leading term. We cannot establish that the correction term in Eq. (7) has the same relevance for the original MAM as we believe the leading-order term does; in the accessible time window of the MAM simulations our data are best described by an effective exponent  $\alpha_{\text{eff}}$ , which reflects a sizeable next-to-leading correction [16]. Current large-scale simulations by Ben-Avraham and Zhong indeed confirm unambiguously both our leading density decay law (1) as well as the power  $t^{-1/2}$  for the first correction term [18].

In this one-dimensional system the reaction zone width  $\ell(t)$  is just equal to the typical interparticle distance between representatives of different species. The reaction rate  $\kappa(t)$  is just the inverse of the time needed to diffuse over this length [17], hence  $\kappa(t) \sim D/\ell(t)^2$ . Combining this with  $\dot{\rho} = -\kappa/L$  and the known time dependences of  $L(t)$  and  $\rho(t)$ , we find  $\ell(t)^2 \sim \rho_0^{-2}(\rho_0^2 Dt)^{\alpha(q)+1/2}$ , whence

with  $\lambda(q) = (2q-1)/4q$ . For  $q = 2$  this reproduces the known result  $\lambda(2) = 3/8$  [17]. The value  $\lambda(\infty) = 1/2$  indicates that for infinitely many species the reaction zones grow as fast as the typical domain size, and hence there can be no segregation. How to aptly take into account the special one-dimensional topological restrictions in a field-theoretic description remains an open issue.

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